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Relationship between column aerosol optical thickness and in situ ground based dust concentrations over Barbados

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Abstract. Aerosol optical depth measurements over Barbados acquired through the AERONET network are analyzed. Optical depth shows a pronounced seasonal pattern, with a maximum observed during summer. The temporal trends of the sunphotometry data were similar to those of ground-based dust concentration measurements. A simple linear regression relationship has been established between mean monthly values of aerosol optical depth and dust concentrations (correlation coefficient r=0.93), which allows the estimation of optical depth for any specific time period when dust concentration measurements are available. The robustness of the regression was validated with an independent data set.

Introduction

Knowledge of aerosol characteristics on a global scale, their temporal change and interrelations with other atmospheric parameters is of great importance if we are to understand the mechanisms which define the aerosol optical state of the atmosphere. One of the major goals of the Global Aerosol Climatology Project (GACP) is a systematic application of the aerosol retrieval algorithms to the whole period of available satellite measurements [*Curran et al.*, 1998]. Retrieved aerosol optical parameters should be validated employing ground-based remote and/or in-situ measurements. Synergism of different data sets and measured parameters might be used to extract aerosol climatology information from existing satellite and other available data. Improved aerosol climatology in turn enables more accurate estimations of the direct and indirect aerosol forcing [*Kaufman et al.*, 1997].

Here we present the results of aerosol optical depth measurements over Barbados, compare optical data in the total atmospheric column with ground-based dust concentration measurements, and investigate the relationship between them.

Data Collection

An automatic Sun and sky scanning radiometer CIMEL was established as an Aerosol Robotic Network (AERONET) site on Barbados in 1996 and made measurements of the direct sun and diffuse sky radiances within the spectral range 440-1020 nm. The CIMEL radiometer was located on the roof of the Meteorological Institute on the western side of the island and several miles north of the capital. A series of papers describe the instrument itself, corresponding measurement sequences and its accuracy [*Holben et al.*, 1998; *Eck et al.*, 1999; *Schmid et al.*, 1999]. Aerosol optical depth data in this paper were cloud screened according to the procedure described in [*Smirnov et al.*, 2000] and quality assured according to the following analysis.

Rapid filter degradation in the harsh environmental conditions on Barbados (sea salt and dust mixture, high humidity) was a problem during the measurement period. However, we found certain channels whose stability and performance were satisfactory. Table 1 presents the deployment and calibration history. Throughout the whole measurement period the 440 nm channel as judged from the calibration history was reliable except for 6 months in 1998. The 870 nm channel was considered to be reliable on 469 days. During 1996 and 9 months of 1997 (217 days) data for this channel was flagged as unreliable. To estimate τ_a(870 nm) during this period we computed the Angstrom wavelength exponent using the 440 nm and 670 nm optical depth pair and extrapolated the aerosol optical depth to 870 nm. In order to verify this procedure we compared measured and calculated τ_a(870 nm) for the period when optical depths in three spectral channels were available. Altogether we found 93 such days (75 days in 1998 and 19 in 1999). Daily averages were consistent within 0.01, except for 4 days, when the difference was 0.02. Despite all the calibration and instrumental problems we believe that the optical depth data which we employed in this study are accurate to within ~±0.02.

Previous results by *Li et al.* [1996] indicate that aerosol scattering and its variance at Barbados is usually dominated by Saharan dust; hence, we concentrate on relationships to this aerosol component. Prospero and co-workers have been sampling airborne particles on Barbados since the late 1960s [*Prospero and Nees*, 1986]. For consistency within the current data analysis, we consider only the samples collected after mid-May 1984 when a

Table 1. Deployment history of CIMEL sun/sky radiometer on Barbados.

Time period	CIMEL	Reliable channels	Calibration	N
6/5/96–23/10/96	#25	440 and 670 nm	pre-field	5
12/12/96–11/9/97	#25	440 and 670 nm	interpolation	212
28/9/97–17/12/97	#74	440 and 870 nm	pre-field	51
18/12/97–30/3/98	#25	440,670, 870 nm	pre-field	75
18/4/98–21/10/98	#25	870 nm	pre-field	149
22/10/98–25/5/99	#74	440 and 870 nm	pre-field	176
28/5/99–22/6/99	#25	440,670,870 nm	pre-field	18

N is the number of averaged days.

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15-m Al fold over tower was installed at the Ragged point sampling site, a 30-m bluff adjacent to the ocean near the easternmost point of the island. On 17 August 1988, this platform was replaced with a standard AEROCE (Atmosphere/Ocean Chemistry Experiment) 20-m walk-up Al tower. Although the AEROCE tower was somewhat taller, the chemistry of the atmospheric particulates (including sea-salt concentrations) measured at the site remained consistent across the change. All samples considered in this report are high volume bulk filter samples collected by drawing air through 20x25 cm Whatman-41 filters at a flow rate of 45-50 m^3 hr^{-1}. The samples are first extracted with deionized water to remove the soluble components ***[Prospero***, 1986, 1999; ***Savoie***, 1984; ***Savoie et al.,*** 1989, 19921. To estimate the dust concentration, the insoluble material is ashed overnight at 500°C. The weight of the remnant material minus that of the average blank is referred to as the ash weight. The dust mass is estimated by multiplying the ash weight by 1.3. The multiplier is based on an Al/ash ratio of 0.104 measured in 997 concurrent samples with ash concentrations greater than 1 $\mu g\ m^{-3}$ at Barbados from 1988 through 1992 and on an assumed Al concentration of 8% in unaltered Saharan dust.

The results from the previous studies cited above and others ***[Chiapello et al.,*** 1995, 1999; ***Prospero,*** 1999; ***Prospero and Carlson***, 1972] have shown that the primary transport of dust from Africa to Barbados takes place in the mid-troposphere in a layer referred to as the Saharan Air Layer (SAL). Transport down into the marine boundary layer (MBL) is largely a consequence of vertical mixing across the trade-wind inversion. Seasonally, the highest mean dust concentration occurs during summer when transport to Barbados is especially persistent and the lowest mean concentration occur during winter when dust events are least frequent. Although dust occurrences are sporadic during the winter and spring months, the sharp peaks in the daily averaged dust concentrations, particularly during spring, are frequently higher than those that occur during the summer.

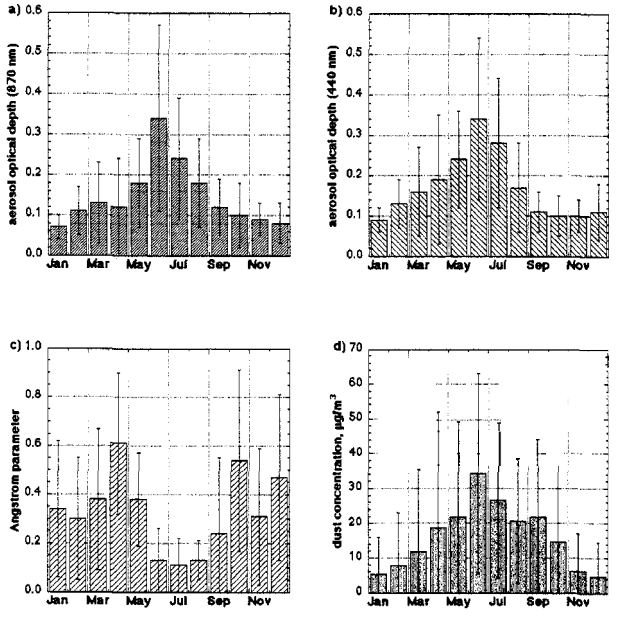

Figure 1. Mean monthly values of aerosol optical depth at the wavelength 870 nm (a), at the wavelength 440 nm (b), and Angstrom parameter (c) for the whole period of optical measurements; mean monthly dust concentrations ($\mu g/m^3$) on Barbados for the period of May 1984 - June 1998 (d) (the bars indicate plus or minus one standard deviation).

Table 2. Number of averaged days for optical and in-situ aerosol measurements.

	1996-1999	1984-1998	τ_a(870 nm), and M, $\mu g/m^3$		
	τ_a(870 nm)	M, $\mu g/m^3$	1997	1998	1999
January	**78**	**414**	**26**	**23**	**29**
February	**71**	**378**	**24**	**24**	**22**
March	**67**	**428**	**28**	**21**	18
April	**43**	**408**	**8**	**10**	**25**
May	**77**	**434**	**21**	**24**	**29**
June	**63**	**440**	**28**	**21**	**14**
July	**52**	**421**	**25**	**27**	
August	**51**	**390**	**23**	**28**	
September	**37**	**344**	11	**22**	
October	**35**	**367**	**14**	**20**	
November	**46**	**389**	**22**	**23**	
December	**65**	**415**	**20**	**26**	

Results

Multi-annual statistics of aerosol optical depth at 870 nm is presented in Figure 1a. A total of 685 daily averages were analyzed (Table 2). The aerosol optical depth shows an increase from May through August, which peaks during June and July. Standard deviations generally increase with the mean values of τ_a. The coefficients of variation (standard deviation over mean) are of the order of 50% or greater. The intra-annual variability of optical depth is consistent with the dust concentration measurements reported by ***Chiapello et al.*** [1995] and ***Prospero*** [1999]. Statistics for the 440 nm channel **look** similar to the 870 nm statistics (Figure 1b), there being fewer measurements (6 months from April till October 1998 were excluded). The Angstrom parameter a, estimated using pairs of wavelengths (440 and 670 nm or 440 and 870 nm), showed that coarse particles (small a) always influence atmospheric aerosol optical properties above Barbados (Figure 1c). The values of α which are typically less than 0.5 indicate the presence of either dust or maritime aerosol or both. It may be observed in Figure 1a-1c that much more dust is present in June, July and August than in any other month (a is below 0.1).

The seasonal cycle of dust concentration over Barbados is presented in Figure 1d based on daily averages for the measurement period from 1984 to 1998. The total number of averaged days for each month is listed in Table 2. The summer maximum is in evidence and the mean monthly values are consistent with the reported results for Barbados (for different time periods ***[Chiapello et al.,*** 1995; Li et al., 1996; ***Prospero,*** 1999]).

Daily average values of τ_a(870 nm) for all years show large day to day variation with a summer peak evident (Figure 2a). The frequency histogram of τ_a(870 nm) is rather narrow; almost 60% of all values are within the 0.05-0.15 range (Figure 2b). The Angstrom parameter frequency distribution (Figure 2c) shows relatively a neutral spectral dependence of optical depth (modal value of α~0.2), which corresponds to large (>1 μm) average particle sizes. Almost 75% of the α values are located within a range of 0.0-0.6.

The overall statistics (1984-1998) differs from the 18 months matching period (January 1997 – June 1998), when the aerosol records from Barbados (dust concentrations) could be considered conjointly with the optical data. For this "test-bed" period we considered only matching days, i.e. when both optical data and aerosol concentrations were available. Figure 3a demonstrates mean monthly dust concentrations for the time coincident data and

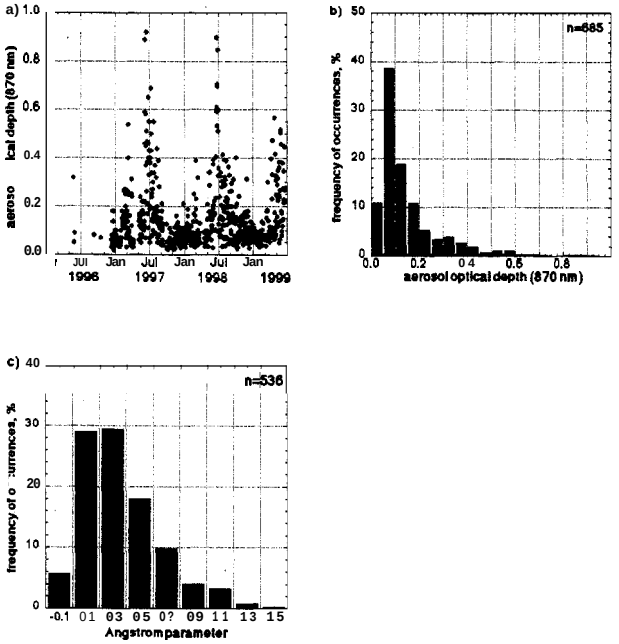

Figure 2. Mean daily values of aerosol optical depth at 870 nm (a), frequency of occurrence of aerosol optical depth (b) and Angstrom parameter (c).

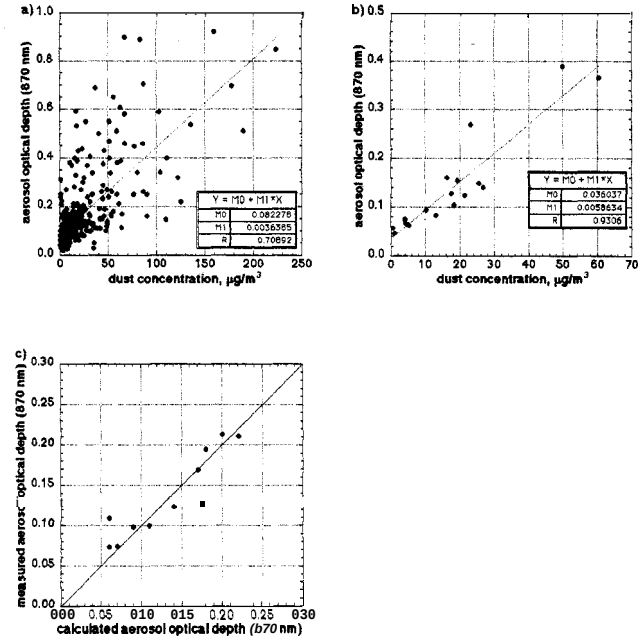

Figure 4. Scattergram of mean daily values (a) and mean monthly values of aerosol optical depth versus dust concentration for the period of January 1997 – June 1998 (b), and scattergram of the measured versus calculated aerosol optical depth for the period of July 1998 – May 1999 (c).

Figure 3b shows aerosol optical depth at a wavelength of 870 nm. A notable correlation is evident. Mean monthly values of the Angstrom parameter are presented in Figure 3c for the 15 month period from January 1997 to March 1998.

A scattergram of daily average aerosol optical depths at 870 nm versus daily dust concentrations shows a lot of scatter (Figure 4a). This scatter can be attributed to various factors. First, averaging time is 24 hours for dust concentration measurements but limited to the daytime for sunphotometer measurements. The second and most important factor is that because of the wind shear between the two layers (MBL and SAL), the peaks in dust concentration in the boundary layer at Barbados are not necessarily coincident with those in the free troposphere immediately above. Moreover, the relative concentrations in the two layers is also dependent on the strength of the vertical mixing between the two layers which is likely to vary significantly depending on the meteorological conditions (e.g. vertical stability) at and upwind of Barbados. As data are averaged over longer time scales, these parameters tend to average to more nearly constant values. As a consequence, the scatter in the relationship decreases substantially and the correlation coefficient increases markedly.

The relationship between monthly averages of dust concentration and aerosol optical depth at 870 nm (Figure 4b) shows strong correlation (correlation coefficient of 0.93). Consideration of one, three and seven day averages yielded weaker correlation (correlation coefficients of 0.71, 0.77 and 0.81 correspondingly). It should be noted that the regression in Figure 4b yielded a non-zero intercept with a value of about 0.04. This is due to the effects of sea-salt and non-seasalt sulfates in the MBL and background aerosol in the free troposphere (FT). This value is consistent with the intercept of 0.015 km^{-1}, reported by *Li et al.* [1996], if we multiply the aerosol scattering coefficient by a

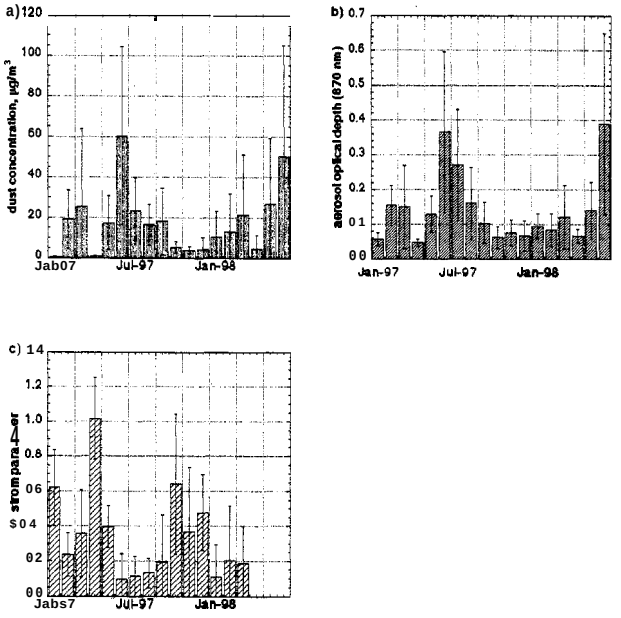

Figure 3. Mean monthly dust concentrations (a), aerosol optical depth at 870 nm (b), and Angstrom parameter (c) for the period of January 1997 – June 1998.

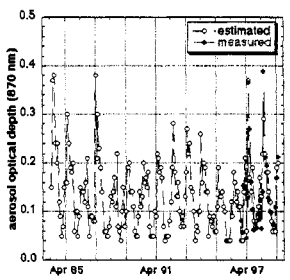

Figure 5. Time series of estimated and measured mean monthly values of aerosol optical depth over Barbados.

Table 3. Regression Parameters and Errors

	1/97-6/98	1/97-12/98	1/97-5/99
Intercept (MO)	0.0362	0.0350	0.0406
Slope (M1j	0.0059	0.0058	0.0056
Standard Error (M0)	0.0137	0.0115	0.0098
Standard Error (MI)	0.0006	0.0005	0.0004

nominal aerosol scale height value (taken as 2 or 3 km, for example).

Additional measurements were considered in order to independently validate the linear relationship between τ_a(870 nm) and dust concentration shown in Figure 4b. Matching dust and optical data for the 11 month period between July 1998 and May 1999, permitted the estimation of τ_a(870 nm) from mean monthly dust concentrations. A scattergram of the measured versus calculated optical depth is presented in Figure 4c. The consistency between measured and estimated τ_a is evident with a root mean square error for these 11 points of 0.023 in τ_a(870 nm), and with the exception of two points, the absolute error was not higher than 0.02.

Using the linear regression shown in Figure 4b we estimated monthly averages of τ_a(870 nm) corresponding to the long-term record of dust concentrations over Barbados. The results are plotted in Figure **5** for the period starting in May 1984 and ending in May 1999 along with the measured mean monthly values. The agreement between measured and estimated τ_a is notable.

Conclusion

A derived relationship between aerosol optical depth and dust concentration allowed the estimation of optical depth for any time period when dust concentration measurements were available. The robustness of the regression can be seen in Table 3, where intercept and slope are presented along with their standard errors for the various time periods. The regression coefficients are stable and errors are small. Currently work is under way to determine similar relationships for other sites where AERONET and AEROCE/NSF networks have reliable long-term records.

The slope of aerosol optical depth versus dust concentration presented here for January 1997 to May 1999 (0.0056 $m^3/\mu g$), together with the slope of total aerosol scattering versus dust concentration, as determined near the surface by *Li et al.* [1996] (0.83 m^2/g), can be used to estimate the dust partitioning between the marine boundary layer and the free troposphere over Barbados. The ratio of these slopes yields a dust (and associated pollutants) scale height of 7 km. Here the scale height is defined as the height of a vertical column **of** constant dust concentration (equal to that which is measured at the surface) that would be required to yield the aerosol optical depth appropriate to the near-surface concentration. For the average marine boundary layer depth of 1 km in this region, the 7-km scale height requires that the column loading of dust in the free troposphere be, on average, about 6 times that in the underlying MBL. Obviously, a wide variety of scenarios of dust vertical distribution could provide this 6 to 1 average ratio and, in fact, the actual vertical distribution is likely to vary substantially. During short-term aircraft flights near Barbados, *Talbot et al.* [1986] found strong layering of the dust and its associated pollutants in the FT. However, when averaged over monthly periods, the sharp layering is likely to be smoothed considerably. Notably, the simple scenario of a 1 km thick MBL with a given dust concentration overlain by a 2 km thick Saharan Air Layer (SAL) with a factor of three higher dust concentration is consistent with the above requirement of a 6:1 ratio as well as the results and conceptual model suggested by ***Prospero and Carlson*** [1972; 1980].

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